

# THERMOCAPILLARY MIGRATION AND INTERACTIONS OF BUBBLES AND DROPS

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## ABSTRACT

Results from ground-based theoretical and experimental research on the motion of bubbles and drops in a temperature gradient are described and a brief account is given of plans for a flight experiment scheduled in 1994.

## INTRODUCTION

When a drop or bubble is placed in another fluid and subjected to the action of a temperature gradient, the drop will move. Such motion is a direct consequence of the variation of interfacial tension with temperature which leads to a thermocapillary stress at the interface between the drop and the continuous phase. Because of this stress, the state of rest in the two fluids adjoining the interface is not tenable. The ensuing motion of the fluids usually leads to a propulsion of the drop, labelled "thermocapillary migration," in the general direction of the warmer fluid. A variation of the composition of species on the interface also can lead to such capillary stresses and drop movement. However, a uniform temperature gradient is more straightforward to establish and maintain when compared to a similar composition gradient. Therefore, virtually all existing experimental research on this subject has focused on thermocapillary migration. The literature on both experimental and theoretical research in this field upto approximately 1989 has been adequately reviewed by Wozniak *et al.* [1] and Subramanian [2].

The movement of suspended objects such as drops and bubbles is relevant to situations that are likely to arise in low gravity experiments. Liquid drops might be encountered during the formation and solidification of alloys, and in separation processes such as extraction that might be used in long duration space voyages for recycling purposes. Also, a dispersion of vapor bubbles might be encountered in heat transfer fluids used in spacecraft which undergo phase change. Gas bubbles may be encountered in crystallization where dissolved gases are rejected at the interface and also in separation processes such as gas absorption. In most applications, it is likely that a collection of drops or bubbles would be involved in which the individual members will influence the motion of each other, and also possibly coalesce leading to changes in size distributions over time.

## RATIONALE FOR EXPERIMENTS

The objective of this research effort is to develop an understanding of the thermocapillary migration and interactions of drops and bubbles from first principles. Both theoretical predictions and experimental observations are necessary to achieve this objective. Predictions from idealized models alone are inadequate for the following reasons.

1. The interface between two fluids is sometimes contaminated by substances which are surface active in the system. Such materials, commonly called surfactants, are specific to each pair of fluids. They adsorb on the interface because this lowers the free energy of the system. However, because of motion along the interface, it is possible to develop and sustain gradients of surfactant

concentration on the interface. This produces a capillary stress which generally tends to oppose that due to the applied temperature gradient. Because of the difficulty in measuring the relevant physical properties of surfactant systems such as adsorption constants and surface diffusion coefficients, it is very difficult to construct precise models of the behavior of systems containing surfactants even when the surface active substance is a known entity. More commonly, trace amounts of a variety of impurities in the chemicals can act as surfactants and this makes the task of modeling even more onerous. Using a simplistic model, Nallani and Subramanian [3] have had some success in describing the behavior of one system (methanol drops in silicone oil) which appeared to contain surfactants. However, this cannot always be done.

2. Even when the system is clean, the fluid in which the drop is submerged is enclosed in a container. In principle, unsteady interactions of the migrating drops with container walls can be modeled even permitting significant convective transport effects and shape deformation, but the computational problem can be substantial. It is useful to obtain experimental data so that the correctness of predictions from models can be independently verified. For similar reasons, experimental data needs to be acquired on pairs of drops. Such pairwise interactions are usually used in modeling systems of multiple drops.

In view of the above, we have been conducting an experimental program on the ground for over a decade to study aspects of thermocapillary migration that one can examine while the system is subject to the earth's gravitational field. Since free convection in the continuous phase is a potential source of problems in interpreting the data, vertical temperature gradients are best to use. The presence of buoyant convection and gravity driven motion of the suspended bubbles or drops places severe restrictions on the length scales of systems that can be studied. This limits the values of two important dimensionless groups that influence the speed of the drops, namely the Reynolds number and the Marangoni number, that can be obtained in ground-based experiments. The Marangoni number is a Péclet number defined using a reference velocity that is appropriate for thermocapillary motion. In addition to the speed of the objects, their shapes are of interest. In this case, the Capillary number plays a significant role in low Reynolds number problems in affecting the shapes of drops and hence their speed, and the Weber number would play a similar role when inertia is important. To conserve space, definitions are not given here, but they are well-known and can be obtained from textbooks on fluid mechanics.

## **IML-2 FLIGHT EXPERIMENTS**

Due to the restrictions introduced by gravity, we are planning to conduct experiments in summer 1994 on the IML-2 mission of the Space Shuttle. The experiments will be performed in the Bubble, Drop, and Particle Unit (BDPU) which is an apparatus designed and built in Europe under the auspices of the European Space Agency. Our plans are to establish temperature gradients in a silicone oil contained in a rectangular cell which is 60 mm long and 45 x 45 mm in cross section. In separate cells, gas bubbles and liquid drops will be injected and their motion photographed. The radii of the bubbles as well as the temperature gradients used will be varied systematically in the experiments so as to cover as wide a range of the Marangoni number as possible. The Reynolds number will be on the order of unity in these experiments. Predictions of the behavior of spherical bubbles for a wide range of Marangoni and Reynolds numbers have been given by Balasubramaniam and Lavery [4]. Also, recently we have obtained asymptotic results for the limit of large values of the Marangoni number both in the low Reynolds number limit and in the limit of potential flow [5, 6]. These predictions will be tested in the IML-2 experiments. Here, we present some recent results from the ground-based research and point out certain interesting aspects of interaction problems.

## GROUND-BASED RESEARCH

### Experimental Apparatus and Procedure

The ground-based experiments were conducted in a test cell schematically shown in Figure 1. The heart of the apparatus is a rectangular region bounded above and below by two aluminum blocks which can be maintained at desired temperatures, thus establishing a vertical temperature gradient in the fluid contained in the enclosure. The side walls are made of teflon and equipped with double glass windows for viewing and lighting purposes. Septums are mounted in the wall to permit insertion of very fine glass capillaries which can be used to inject small bubbles or drops. The procedure consists of filling the cell with the desired continuous phase liquid and establishing an appropriate temperature gradient, followed by the insertion of one or more bubbles or drops. The entire process of injection and migration is recorded using a video camera mounted on a microscope. The video tape is later analyzed frame by frame to obtain information on drop/bubble sizes and velocities.

### Results and Discussion

Using this test cell, Srividya [7] performed experiments on fluorinert FC-75 drops in a Dow-Corning DC-200 series silicone oil of nominal viscosity 50 cs contained in the test cell. FC-75 is more dense than the silicone oil, and an upward temperature gradient was used so that the thermocapillary effect would retard the downward settling of the drops. Conditions were set such that the drops, in the region of observation, experienced an average temperature of either 20° C or 40° C. Data were obtained on drops ranging in radius from 60 to 150  $\mu\text{m}$  and in upward temperature gradients ranging from 4 to 11 K/mm. In separate experiments, the temperature field in the cell was measured in the region where the drops migrated and was confirmed to be linear. Also, the residual free convection in the cell was characterized in experiments using tracers and the magnitudes were found to be within the measurement uncertainty of the drop velocities. The Reynolds number in the experiments were less than 0.015.

The data obtained at an average temperature of 40°C are shown in Figure 2. As anticipated, the drops settled at velocities predicted by the standard Hadamard-Rybczynski model for low Reynolds number motion under isothermal conditions. Under the action of an upward temperature gradient, they settled at lower speeds, with the overall reduction being enhanced for larger temperature gradients. The data collapse onto a single straight line when the thermocapillary contribution to the velocity is extracted and plotted against the product of the applied temperature gradient and radius. The slope of this straight line can be used to infer the rate of change of the interfacial tension with temperature which was determined to be  $-0.0397\text{mN}/(\text{m}\cdot\text{K})$ . This value is somewhat higher than the value of  $-0.03041\text{mN}/(\text{m}\cdot\text{K})$  obtained from a series of interfacial tension measurements made at different temperatures ranging from 5 – 65°C. However, both values are subject to experimental uncertainty. The static measurements suffer from an inability to maintain perfectly still conditions in the liquids and the actual interfacial tension values are only good to approximately 0.1 mN/m. On the other hand, the migration experiments were performed under conditions wherein the Péclet number was as large as 5. The theory used to fit the data assumes the Péclet number to be negligible.

In another series of experiments in a similar cell [8] pairs of air bubbles were injected into a DC-200 silicone oil of nominal viscosity 1000 cs to observe interaction effects both under isothermal conditions and in the presence of a downward temperature gradient. In the non-isothermal runs, care was taken to maintain the Rayleigh number substantially below the critical value at which cellular convection would be triggered. In separate experiments, the magnitude of the residual convection in the liquid was characterized and found to be well within the uncertainty of the velocities measured. Convective transport of both energy and momentum was negligible in the experiments.

Under these conditions, the results for bubble velocities were found to be generally consistent with predictions from theoretical models. Sometimes, the experiments produced counter-intuitive observations; for example, when a large bubble and a small bubble are present in a downward temperature gradient that is sufficient to force the large bubble downward, the small bubble is seen to move upward! If the small bubble were isolated, it would move downward quite rapidly under these conditions. The reason is a substantial upward draft caused by the large bubble even though it is moving downward; this is a consequence of the Stokeslet contribution to the flow around this bubble. The velocity field around an isolated bubble moving under these conditions is a superposition of a Stokeslet and a potential dipole from the gravity and thermocapillary contributions respectively.

While Fluorinert-silicone oil form a suitable candidate pair for flight experiments, the large density difference between them precludes interaction experiments on the ground under conditions when convective energy transport is important. Therefore, we have identified the pair diethyl maleate - propanediol as a candidate for ground-based experiments. Preliminary experiments on isolated drops show confirmation of the expected scalings at negligible Reynolds and Péclet number. Experiments on pairs are planned in the coming year.

Wei and Subramanian [9] have solved the problem of multi-bubble interactions under conditions of negligible Reynolds and Péclet number using the method of boundary collocation. Space limitations prohibit us from presenting these results in full detail here, and only interesting samples are provided. In Figure 3 the streamlines in a laboratory reference frame are displayed for motion induced by two bubbles which are migrating in a large body of fluid. The flow patterns corresponding to the thermocapillary migration problem are contrasted with those for body force driven motion. The differences are striking. In the axisymmetric case, a separated reverse flow wake is observed. By reversing the order of the large and small bubbles, it is possible to produce a flow pattern in which this reverse flow region occurs in front of the migrating pair. In the asymmetric case (wherein the line of centers is normal to the temperature gradient), to the right of the migrating bubble pair, fluid from far away is drawn toward the bubbles and thrown outward. These interesting flow patterns arise from the nature of the driving force in thermocapillary migration which is the temperature gradient field on the surfaces of the bubbles. In the case of an isolated bubble under similar conditions, the temperature field, arising from the solution of Laplace's equation in the fluid, is the first Legendre Polynomial in the cosine of the polar angle. The resulting flow is a potential dipole which possesses fore-aft symmetry. When higher Legendre modes are excited by breaking the symmetry of this situation, complex flow patterns arise as a consequence of the superposition of relatively simple multicellular flows driven by the pure Legendre modes. Additional discussion can be found in [9].

Results for the velocities of the individual bubbles in a pair were calculated by the boundary collocation method and compared with predictions from the method of reflections. Also, limited results were calculated for a chain of three bubbles whose centers lie on a straight line which is either parallel or perpendicular to the applied temperature gradient. In this case, a pairwise-additive approximation using the reflections solution was constructed. The approximations perform very well when the bubbles are far apart, but begin to depart from the correct solution when the distance of separation between the bubble surfaces is less than approximately one-half the radius of the larger bubble in the pair. Further details are given in [9].

It is important to emphasize that the above calculations in the case of more than one bubble are limited to the linear problem wherein convective transport effects are neglected altogether and several other simplifying assumptions are made. There is a need for theoretical predictions to be made

permitting significant convective transport effects and also accommodating shape deformations.

### CONCLUDING REMARKS

In the future, this research will continue in the direction of experiments on systems involving more than one drop (or bubble) interacting with each other and possibly with the boundaries of the system. A good beginning would be to work with a pair of such objects introduced at appropriate well-defined locations in the test cell after a temperature gradient has been established. On the ground, one is restricted to investigating a very small range of parameters in the thermocapillary migration problem because of the complicating effects of gravity on the motion of the drops. Therefore, it will be necessary to perform experiments under reduced gravity conditions in order to fully explore a good range of parameters in this problem. It would be more difficult to perform studies of the actual approach and coalescence of drops since coalescence is usually a very rapid process. However, it is envisioned that as the study matures, attempts to explore coalescence will be undertaken.

### ACKNOWLEDGMENTS

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### References

- [1] G. Wozniak, J. Siekmann, and J. Srulijes, *Z. Flugwiss. Weltraumforsch.*, **12** (1988) 137-144.
- [2] R.S. Subramanian, "The motion of bubbles and drops in reduced gravity," in *Transport Processes in Drops, Bubbles, and Particles*, (R. Chhabra and D. Dekee, Eds.), Hemisphere, 1992, pp. 1-41.
- [3] M. Nallani and R.S. Subramanian, *J. Colloid Interface Sci.*, **157** (1993) 24-31.
- [4] R. Balasubramaniam and J.E. Lavery, *Numerical Heat Transfer A*, **16** (1989), 175-187.
- [5] R. Balasubramaniam, "Thermocapillary Bubble Migration - Solution of the Energy Equation for Potential-Flow Approximated Velocity Fields," International Association for Mathematics and Computers in Simulation Symposium, Bangalore, India, December 1992.
- [6] R. Balasubramaniam and R.S. Subramanian, "Thermocapillary Bubble Migration - Thermal Boundary Layers for Large Marangoni Numbers," submitted for publication, *Journal of Fluid Mechanics*, August 1993.
- [7] C.V. Srividya, "Migration of Fluorinert FC-75 Drops in a Silicone Oil in a Vertical Temperature Gradient," M.S. Thesis in Chemical Engineering, Clarkson University, 1993.
- [8] H. Wei and R.S. Subramanian, "Interactions between two bubbles under isothermal conditions and in a downward temperature gradient," submitted to *Phys. Fluids A*, 1994.
- [9] H. Wei and R.S. Subramanian, *Phys. Fluids A*, **5** (1993) 1583-1595.

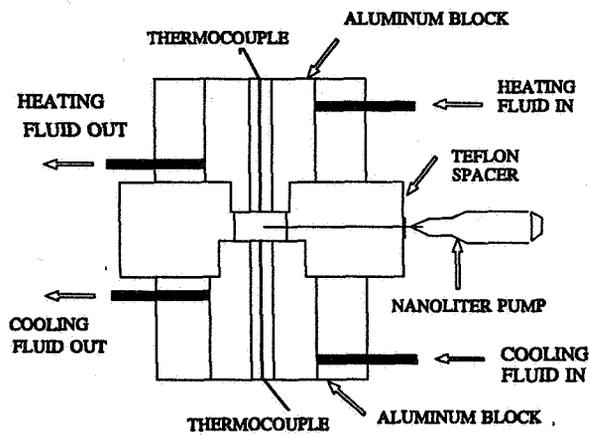


Figure 1. Sketch of Test Cell

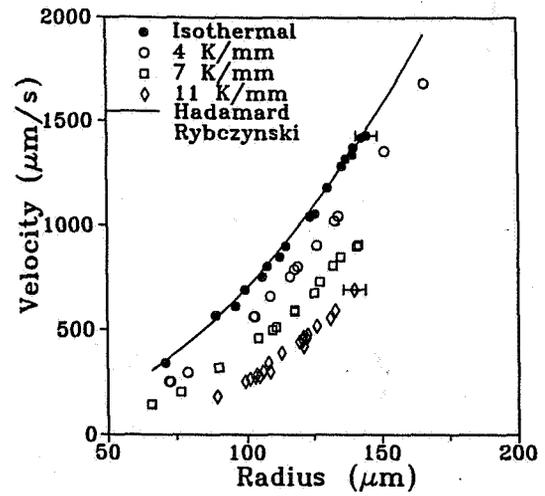
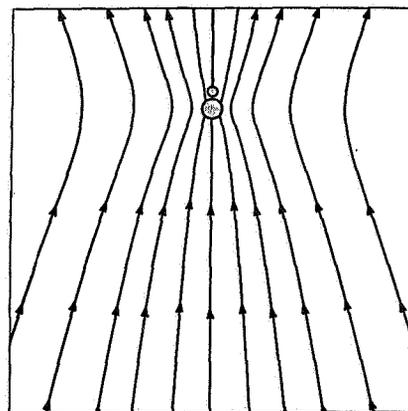
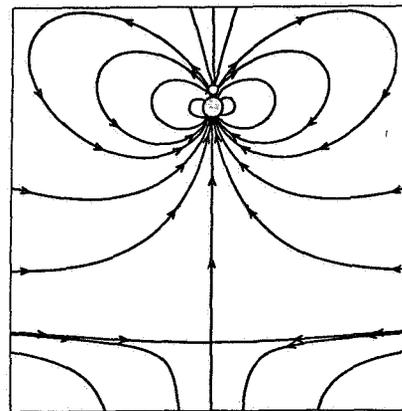


Figure 2. Downward velocity of Fluorinert FC-75 drops in a Dow-Corning DC-200 series silicone oil of nominal viscosity 50 cs

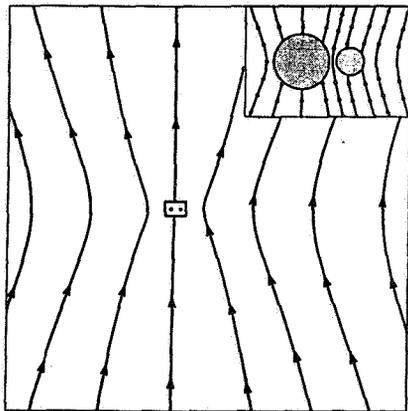


Body Force Driven Motion

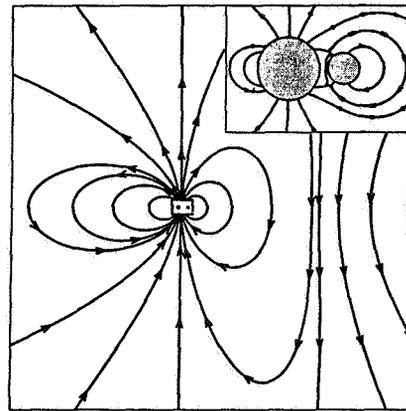


Thermocapillary Migration

(a)



Body Force Driven Motion



Thermocapillary Migration

(b)

Figure 3. Streamlines in a laboratory reference frame for flow generated by a pair of non-identical bubbles undergoing body force driven motion or thermocapillary migration. (a) axisymmetric case - a meridian section is shown. (b) asymmetric case - a section in a symmetry plane is shown. Velocity normal to this plane is zero.